

Constraining kinetic parameters of gas particle interactions in the transition from liquid to solid

Markus Ammann^{1,*}, Sarah Steimer¹, Ming-Tao Lee¹, Goran Grzinic¹, Manabu Shiraiwa², Ulrich Pöschl², Andrew J. Huisman³, Ulrich K. Krieger³, Thomas Peter³, Thomas Berkemeier⁴, and Thomas Koop⁴

¹ Paul Scherrer Institut, Villigen, Switzerland

² Max Planck Institute for Chemistry, Mainz, Germany

³ Institute for Atmospheric and Climate Sciences, ETH, Zürich, Switzerland

⁴ Bielefeld University, Bielefeld, Germany

* Corresponding author: Markus.Ammann@psi.ch

Reactive uptake experiments provide a means to understand aerosol aging in the atmosphere. Kinetic limitations in gas particle interactions are not only playing a role in trace gas budgets and particle processing by oxidants, but also in aerosol formation and growth. The recognition of temperature and humidity mediated viscosity changes for organic solutes and their potential effects on chemical and physical processes governing aerosol evolution also in the troposphere have led to a revived interest in better understanding the mechanisms and kinetics of gas-particle interactions (1,2). We have performed uptake experiments of O₃ and nitrogen oxides to a number of aqueous organic and inorganic or mixed composition substrates to constrain the kinetics of these systems under a wider range of physical conditions, i.e., solid, semi-solid, and liquid, by means of surface accommodation, surface reactions, surface to bulk transfer, bulk diffusion and reaction (3-5). Previous and new data are reanalyzed by fitting with a kinetic flux model (6) that allows to explicitly track non-steady state conditions especially within the particle bulk to account for diffusion of the reactants. Some of the issues related to constrain kinetic parameters and to differentiate surface from bulk processes based on datasets typically limited in coverage of parameter space are discussed.

References

- (1) Koop, T.; Bookhold, J.; Shiraiwa, M.; Pöschl, U. *Phys. Chem. Chem. Phys.*, **2011**, 13,
- (2) Shiraiwa, M.; Ammann, M.; Koop, T.; Pöschl, U. *Proc. Natl. Acad. Sci.* **2012**, 108, 11003-11008.
- (3) Rouvière, A.; Sosedova, Y.; Ammann, M. *J. Phys. Chem. A*, **2010**, 114, 7085-7093.
- (4) Sosedova, Y.; Rouvière, A.; Gäggeler, H. W. Ammann, M. *J. Phys. Chem. A*, **2009**, 113, 10979-10987.
- (5) Sosedova Y.; Rouviere A.; Bartels-Rausch T.; Ammann M. *Photochem. Photobiol. Sci.*, 2011.
- (6) Shiraiwa, M.; Pfrang, C.; Poeschl, U. *Atmos. Chem. Phys.*, **2010**, 10, 3673-3691. 19238-19255.